Ion Mobility Discontinuities in Superfluid Helium: A Test of the Huang-Olinto Theory*

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A new method has been developed for making sensitive differential measurements of ion mobilities in liquid helium. Using this method, it has been possible to make a definitive test of the part of the Huang-Olinto theory intended to explain discontinuities in ion mobilities in superfluid helium. The theory has been found to be incorrect.

I. INTRODUCTION

IN the time since the discovery of quantized ion mobilities in liquid helium, hereafter called "steps," was first announced by Careri *et al.* in 1961,[1] much experimental work has been done and a number of theoretical models[2–4] have been proposed to explain the phenomenon. Of these, the most comprehensive, and perhaps the most ingenious, is that of Huang and Olinto.[4] In this paper it is demonstrated that that part of the Huang-Olinto theory that deals specifically with the steps is not correct.

In order to test the relevant part of the Huang-Olinto theory, a new method has been evolved for making sensitive differential measurements of ion mobilities with respect to various parameters. This method will be described below.

The mobility of an ion is defined by the relationship

$$v=\mu\mathcal{E}, \qquad (1)$$

where μ is the mobility, v is the velocity of the ion, and $\mathcal{E}$ is the electric field acting on the ion. Thus, if the drag force acting on the ion is simply proportional to its velocity, the mobility is a constant with respect to v and $\mathcal{E}$. Careri *et al.*[1,5] discovered that in liquid helium at temperatures near 1°K, the mobility is indeed constant up to a velocity of 5.2 m/sec.[6] However, at this critical velocity, and at approximately integral multiples of it, the mobility suffers a small discontinuity, falling to a new constant level on the order of 6% below the previous one [Fig. 1(a)]. The mobility and critical electric fields are temperature-dependent, but the critical velocity is constant. After six or seven such steps a new phenomenon sets in, in which the velocity actually decreases with increasing electric fields. The full v-$\mathcal{E}$ spectrum is shown in Fig. 1(b).

It has been shown subsequently that the extra drag force on the ion associated with the steps is due to an extra interaction with the normal fluid component of He II,[7] and that the phenomenon persists up to the λ point (2.17°K), although the first critical velocity ceases to be a constant, taking on a complicated temperature dependence above about 1°K.[8] The Huang-Olinto theory[4] is in clear disagreement with this latter work; however, the theory was not designed to deal with the phenomenon at higher temperatures, where the quasiparticle picture of He II breaks down.

For purposes of the present analysis, the theory[4] may be divided into three logically independent parts.

(1) It is postulated that at a velocity given by the relationship 19 $R_iV=nh/m$, where R_i is the ion[9] radius

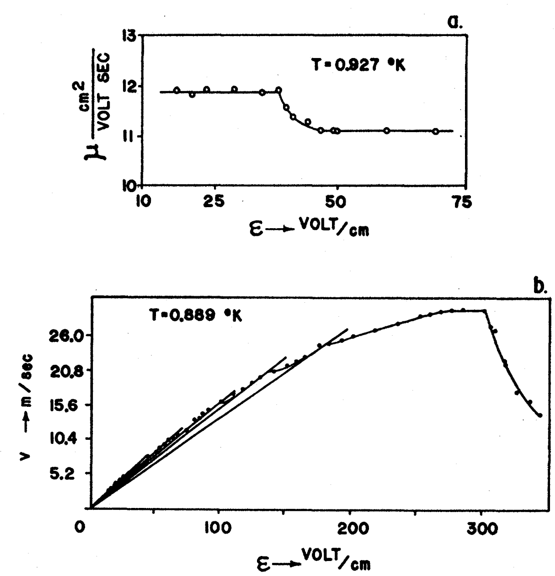

FIG. 1. (a) Example of a mobility step, after G. Careri, S. Cunsolo, and P. Mazzoldi, Phys. Rev. Letters **7**, 151 (1961); (b) the v-$\mathcal{E}$ spectrum after G. Careri, S. Cunsolo, and P. Mazzoldi, Phys. Rev. **136**, A303 (1964).

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[1] G. Careri, S. Cunsolo, and P. Mazzoldi, Phys. Rev. Letters **7**, 151 (1961); see also Ref. 5.

[2] J. A. Cope and P. W. F. Gribbon, in *Proceedings of the Ninth International Conference on Low-Temperature Physics, Columbus, Ohio*, edited by J. A. Daunt *et al.*, (Plenum Press, Inc., New York, 1965) p. 153; Phys. Letters **16**, 128 (1965).

[3] C. Di Castro, Nuovo Cimento **42**, 251 (1966).

[4] K. Huang and A. C. Olinto, Phys. Rev. **139**, A1441 (1965).

[5] G. Careri, S. Cunsolo, and P. Mazzoldi, Phys. Rev. **136**, A303 (1964).

[6] For positive ions; 2.4 m/sec for negative ions.

[7] C. Careri, S. Cunsolo, and M. Vicentini-Missoni, Phys. Rev. **136**, A311 (1964).

[8] L. Bruschi, P. Mazzoldi, and M. Santini, Phys. Rev. **167**, 203 (1968).

[9] Throughout, we use the term "ion" to mean "ion complex" or "charge carrier."

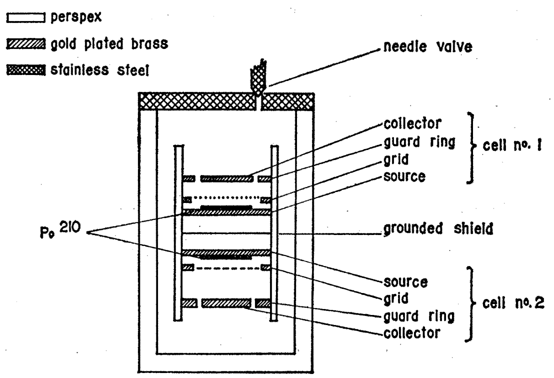

FIG. 2. Experimental double cell, as described in the text.

and h/m is the quantum of circulation, the ion can and does create a quantized vortex ring of circulation nh/m. In order to emphasize the presumably fundamental nature of the relationship, the authors write it as

$$6\pi R_i v = nh/m. \tag{2}$$

The authors suggest a crucial test of the theory, which in fact tests only this postulate. There now exists some experimental evidence both for[10] and against[11] the postulate (at least for $n=1$), but it should be emphasized that the other parts of the theory are independent of it. This part of the theory is not tested in the present experiment.

(2) A recipe is presented for a behavior of the ion which would result in the observed steps: The ion initially accelerates rapidly to the first critical velocity, then remains at that velocity for a fixed time, during which it gathers enough excess energy from the electric field to create a vortex ring at the same velocity. It then sheds the ring and goes to the next critical velocity, and so on, repeating the process until it reached its terminal velocity, determined only by the interaction of the bare ion with the normal fluid. The velocity that one measures experimentally is thus the time average of the various critical velocities and the terminal velocity of the ion, and there results an apparent discontinuity in the mobility after the terminal velocity reaches each critical velocity. The resulting formula makes an excellent fit to the experimental data at $T \approx 1°$K, as may be seen in Figs. 3 and 4 of Ref. 4.

It is this part of the theory which is tested in the present experiment. It follows immediately from the above description that the measured ion velocity depends on the length of the space through which the ions drift. This is because the times spent at the various creation velocities are fixed physically, so that the amount of terminal velocity entering in the average depends on the length of the drift space. Accordingly, the measurement to be presented here is a differential measurement of mobility with respect to the length of the drift space.

(3) The theory goes on to present a criterion for the formation of a stable ion-vortex ring complex. At electric fields higher than that given by this criterion, the ion is trapped by the first vortex ring that it creates and its measured velocity decreases with increasing electric field, a behavior characteristic of the dynamics of vortex rings. There is now considerable evidence that this criterion for a stably charged vortex ring is essentially correct.[12] We shall not concern ourselves with this part of the theory.

II. TECHNIQUE

The method of making differential measurements is based on the technique developed by Cunsolo[13] for measuring ion mobilities. In the Cunsolo technique the ion moves through a drift space under the influence of an electric field in the form of a square wave, averaged to zero. During the positive half-cycle, the ions are acted upon by a force $e\mathcal{E}$, where $\mathcal{E}$ is the amplitude of the square wave; during the negative half-cycle they are swept out of the drift space with the same force. The current collected depends on the duration of the half-cycle; the ions remaining in the drift space when the field switches polarity are swept out. The zero of current occurs when the half-period is just equal to the ion time of flight. A plot of current i versus frequency of the square wave ν is a straight line, which is extrapolated to zero current to find the time of flight of the ions. The ion velocity is then the drift length divided by the time of flight, and the mobility is obtained from Eq. (1).

The differential method employs two complete versions of the standard experimental cell, each consisting of an ionizing source, a grid, and a collector, the drift space being that between the latter two. All electrodes are made of gold-plated brass, Po^{210} is diffused into the surface of the source electrode, and the drift space is confined by appropriate shields to the central portions of the electrodes where the field is uniform. A small dc field between the source and the grid extracts ions of the desired sign. The two cells are contained together in a single open envelope of Perspex, placed within a second sealable Perspex container. This latter is immersed in a liquid-helium bath at the desired temperature, filled with liquid helium, and sealed with a needle valve. The situation is depicted in Fig. 2.

The two cells are driven by independent square waves, their frequencies measured on a Helwett-Packard No. 52452 electronic counter, and their currents measured by a Cary No. 31 vibrating-reed, master-slave electrometer pair.

In principle, the method consists of setting the currents in the two cells equal at zero frequency (first

[10] S. Cunsolo, B. Maraviglia, and M. Ricci, Internal Note No. 146, Institute of Physics, University of Rome, 1967 (unpublished).
[11] L. Meyer, Phys. Rev. **148**, 145 (1966).
[12] G. Careri, S. Cunsolo, P. Mazzoldi, and M. Santini, Phys. Rev. Letters **15**, 392 (1965); see also Ref. 10. *Note added in proof:* Opposing evidence is presented in G. W. Rayfield, Phys. Rev. **168**, 222 (1968).
[13] S. Cunsolo, Nuovo Cimento **21**, 76 (1961).

point) and then measuring the ratio of frequencies at which the two currents are again equal (second point). It follows from the geometry of the curves that this ratio is just the reciprocal of the times of flight in the two cells. The master-slave electrometer pair has a provision for choosing a part of the slave current by means of a precision voltage divider, and subtracting it directly from the master current. This is the technique used for setting the currents equal at the first point; the current measured in these i-ν curves is always arbitrary, and a fixed part of the slave current serves just as well as the whole slave current. The currents are set equal at the second point by varying the frequencies of the square waves, leaving the voltage divider fixed.

Thus the problem is reduced to measuring the ratio of two frequencies; there is no extrapolation to be made, and small temperature changes are unimportant since both cells share the same superfluid and thus change together.

An analysis of the standard Cunsolo technique yields three principal sources of scatter in the results: (1) uncertainty in the extrapolation of the i-ν line to zero current, of about 1%, (2) small changes in temperature during the measurement of a single mobility point; a drift in temperature of 0.001°K introduces a change of approximately 1.5% in the mobility, and (3) uncertainty in the amplitude of the square wave. The magnitude of the uncertainty depends on the method used for making the measurement.

An over-all scatter of 2–3% is normally observed in these measurements. The present technique obviates uncertainties of types (1) and (2), at the expense of making differential (or relative) rather than absolute measurements. Above all, problems normally associated with comparing experimental data from different runs, such as precise reproduceability of the temperature and the condition of the bath, as well as the length of the cell, are not encountered with this method. For the measurement of the field, a technique has been chosen which has a relative uncertainty on the order of 0.5%, and this is probably the major source of error in the experiment.

The method as used in practice is illustrated schematically in Fig. 3. It is not actually possible to set the currents equal at zero frequency (which is not simply related to dc in this case) nor, indeed, even at the same frequency. Instead, one sets the currents equal at two nearly equal low frequencies, ν_{01} and ν_{02}, measures the ratio of higher frequencies f_1 and f_2 at which the currents are again equal, and measures by the usual technique (or takes from published data) the current zeroing frequency in one of the cells, say, ν_2. The inverse ratio of times of flight α is then given by

$$\alpha=\frac{\nu_1}{\nu_2}=\frac{f_1}{f_2}+\frac{(f_1/f_2-1)(1-f_2/\nu_2)}{f_2/\nu_0-1}, \tag{3a}$$

where

$$\nu_0\cong(\nu_{02}-\nu_{01})f_2/(f_1-f_2)+\nu_{02}. \tag{3b}$$

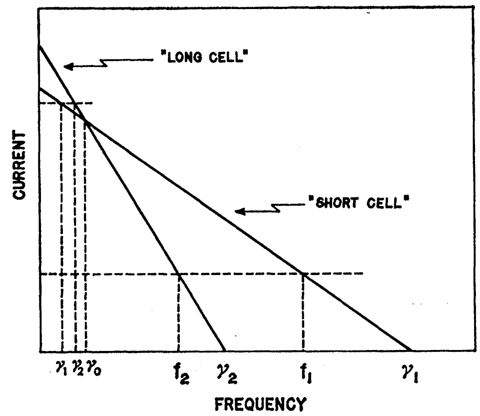

FIG. 3. i-ν curves in the double-cell method. The notation is explained in the text.

This is actually less complicated than it seems, since the correction term in Eq. (3a) is seldom larger than 2% of f_1/f_2, so that errors in ν_0 and ν_2 are of little importance, and the calculation is easily done.

The sensitivity with which one can set the currents equal at the first point (ν_{01} and ν_{02}) may be measured by changing the setting of the voltage divider. It is found that a change of a few parts per thousand in the fraction of the slave current fed back is easily detected, even though the net current (read on a chart recorder) is somewhat noisy at this point due to beating between the two nearly equal frequencies. At the second point (f_1 and f_2) it was often found that an error of one part per thousand in the ratio f_1/f_2 was detectable.

The technique used for measuring the square-wave amplitude is shown in Fig. 4. The capacitor C is charged during the positive half-cycle, but during the negative half-cycle, when the diode D does not conduct, C cannot discharge appreciably through the large resistance R. The meter M measures the dc charge level of the capacitor C, and the deflection thus produced is compared directly to that due to a dc potential supplied by a Keithley-type 241 dc power supply (rated at 0.05%). The two square waves are measured by separate instruments. From relative measurements using both instruments on the same square wave, it is estimated that the ratio of amplitudes is known to better than 0.5%.

At low drift velocities (v below $\sim$5 m/sec) the mobility is expected to be independent of drift length, so

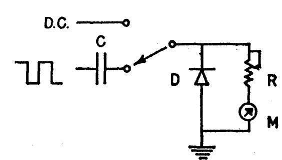

FIG. 4. Device for measuring the amplitude of the square wave, as explained in the text.

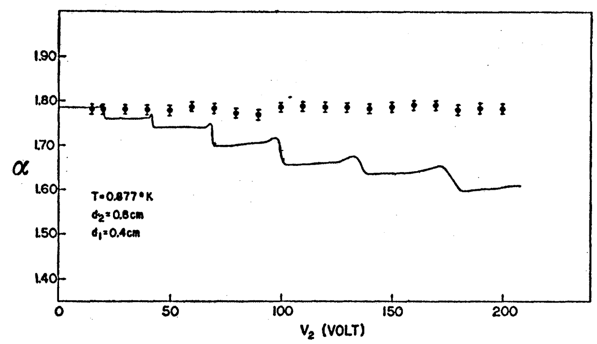

FIG. 5. Experimental results (circles) compared to the predictions of the Huang-Olinto theory (solid line). Error bars on experimental points are ±0.5%.

that the measured ratio α_0 is given by

$$\alpha_0=\frac{\tau_2}{\tau_1}=\frac{\mu^{(1)}}{\mu^{(2)}}\frac{V_1L_2^2}{V_2L_1^2}=\frac{V_1L_2^2}{V_2L_1^2}, \tag{3c}$$

where τ is the time of flight, all subscripts and superscripts refer to the number of the cell, and where $V_i=\mathcal{E}_iL_i$ are the drift potentials. Since the V_i are free to be chosen, these low velocity points may be regarded as a precise measurement of the ratio of drift lengths, L_2/L_1; the ratio of $(L_1/L_2)^2$ is measured to $\frac{1}{2}\%$. It was found that on thermal cycling to room temperature, the ratio of lengths changed by as much as 2%.

The ratio V_2/V_1 was kept constant in each run, and always a few percent different from L_2/L_1, so that the electric fields in the two cells always differed by a few percent. This proved a useful technique and illustrates one of the potential applications of the double-cell method. The mobility is measured by the conventional technique in one of the cells. If two points at adjacent fields show a rapid change in mobility, the ratios α at the two points immediately show whether the change is real or due to experimental scatter. Assuming the normal degree of temperature control, there is no reason why the two cells should both undergo sharp changes in mobility at the same time, when their fields are slightly different. Thus if the change is real, one should find the same change also in α, indicating that the unwatched cell has not changed.

An obvious variation of the technique, using a single square wave suitably divided and measuring the change in the ratio of currents between two frequencies, has also been used successfully, and may be preferable in certain applications.

III. RESULTS AND DISCUSSION

We have measured α, the inverse ratio of times of flight of two experimental cells with different drift lengths. Typical results are shown in Fig. 5, where they are compared to the predictions of the Huang-Olinto theory.[4]

In the theory, the time spent at each of the creation velocities is calculated from the energy required to create the postulated vortex ring; this result, together with the recipe described in Sec. I, leads to the expression

$$v=v_\infty(1+S_n/L)^{-1}, \tag{4}$$

where L is the length of the drift space, and

$$S_n=\frac{E_0}{e\mathcal{E}}\frac{v_\infty}{v_c}\sum_{l=1}^{n}\frac{l(1-lv_c/v_\infty)}{1-\mathcal{E}_l/\mathcal{E}}. \tag{5}$$

Here E_0 is the energy required to make a singly quantized ring at the first critical velocity v_c, $\mathcal{E}_l$ is the electric field at the step, e is the electron charge, and v_∞ is the terminal velocity of the bare ion, which the authors

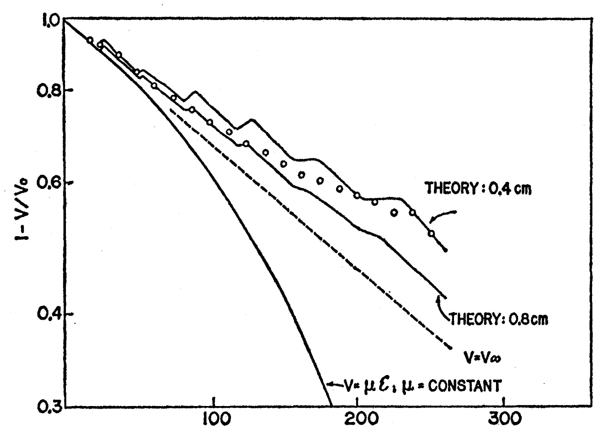

FIG. 6. Plot of $(1-v/v_0)$ versus $\mathcal{E}$. The experimental velocities, measured concurrently with the data of Fig. 5, are compared to curves of μ=const, v_∞, the velocity expected in the Huang-Olinto theory if the mechanism of the steps were absent, and the full Huang-Olinto theory.

interpolate by

$$v_\infty=v_0[1-\exp(-\mathcal{E}/\mathcal{E}_0)], \tag{6}$$

v_0 being the roton creation velocity (about 58 m/sec) and $\mathcal{E}_0=v_0/\mu_0$, where μ_0 is the zero-field mobility.

For the present measurement, the theory predicts

$$\alpha=\alpha_0(1+S_n/L_2)/(1+S_n/L_1), \tag{7}$$

where α_0 is given by Eq. (3c). Equation (7) is not expected to be valid, however, for fields just larger than each critical field. In this case, the final ring is made very slowly and is not completed before the ion reaches the collector. Thus its velocity never reaches v_∞, and Eq. (7) is not correct. Instead, one must use two modified versions of Eq. (7), one for the interval where both cells are too short to complete the final ring, and the other for the interval where only the shorter cell is too short. The result is a somewhat complicated behavior in these intervals.

The predictions of the theory have been calculated for two points between each pair of steps, in the regions where Eq. (7) is expected to be valid, and also at points within the special intervals after the second and sixth steps. The curve indicated by these calculations has been plotted in Fig. 5, where it is compared directly with the experimental results.

It is clear from Fig. 5 that the dependence of ion velocity on drift length which is predicted by the Huang-Olinto model is not present. Since this dependence is central to the model, we shall conclude that, on the basis of these data, the model cannot be correct. First, however, we wish to deal with a possible objection to this conclusion.

In general, it is difficult to detect distinct steps at all after the first few[5] (although, indeed, in certain cases, as many as seven distinct steps have been observed and reported[8]). In the mobilities which were measured separately in the present experiments, distinct discontinuities were never observed past about the third step. It could be argued that, in the absence of steps, a theory intended to account for the steps need not apply, and that the strong disagreement between theory and experiment at high fields in Fig. 5 is not a valid test of the theory.

However, the theory predicts a gross dependence of the ion velocity on electric field, with the effective drag forces being due both to the interaction of the bare ion with the normal fluid, and to the production of vortex rings which leads to steps. Even if the fine structure is smeared out, so long as the velocity obeys the gross dependence, the theory would call for a gross dependence of drift velocity on drift length; to be sure, the fine structure of this dependence would also be smeared out, but the amplitude should be essentially unaffected. In other words, the theory is fundamentally a model to account for the departure of the ion velocity from v_∞ of Eq. (6). So long as the measured velocities do depart from v_∞ in the manner predicted by the theory, the length dependence must also be present.

Thus, in order to be certain that the wide divergence between theory and experiment shown in Fig. 5 is meaningful, we must show that v deviates from v_∞ in the way called for by the theory. Figure 6 is a semilogarithmic plot of $(1-v/v_0)$ against $\mathcal{E}$. The dotted line is for $v=v_\infty$. The theory predicts departures from v_∞ shown by the solid curves for cells of 0.4- and 0.8-cm drift spaces. Superimposed are the measured values which we have already shown to be independent of length. Clearly, the gross departure from v_∞ that the theory is intended to account for is present. We may therefore conclude that the difference between theory and experiment at high fields in Fig. 5 is indeed significant.

The above argument becomes much simpler, of course, if one can show a clear divergence between theory and experiment when the steps are observable. However the steps are most easily observable just where the predicted dependence of velocity on drift length is smallest, in the first few steps. Nevertheless, an example of the required behavior is shown in Fig. 7. Here a second step has been chosen because the step is still reasonably distinct, whereas the predicted effect is already outside of the uncertainties of the experiment. Figure 7 shows the ratio α, the prediction of the theory, and the mobilities μ. Once again, we conclude that the theory is not correct.

In fact, the simplicity of the present result, $\alpha=\text{const}$, implies that it should be a transparent feature of any successful theory.

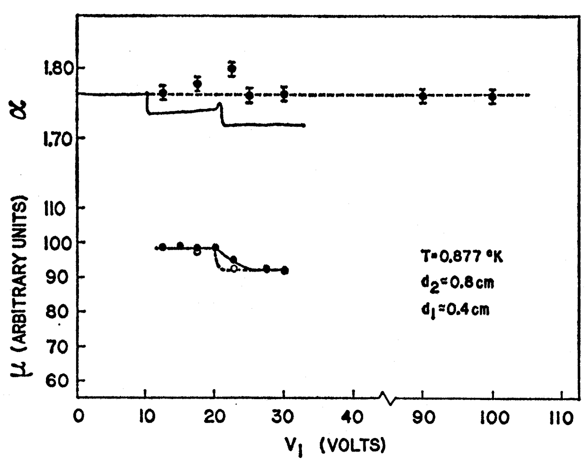

FIG. 7. Example of a second step in the mobility, together with the measured ratio α, and the prediction of the theory for α. Note the point outside of experimental error in α. It indicates that the step was clean (had already reached the lower level) in the unwatched cell. The behavior of the unwatched cell, where it deviates from the watched cell, is indicated by the open circles and the dashed line.

IV. CONCLUSION

From the beginning, it seemed an attractive idea to account for the steps as due to quantized vortex rings. It has now been convincingly demonstrated that quantized vortex rings do exist and interact with ions in He II,[14] and this discovery has given fresh impetus to the idea of somehow accounting for the steps by means of vortex rings. In the present work we have shown that the most significant attempt thus far to do this is not correct.

Nevertheless, critical-velocity phenomena in He II have long been explained in terms of vortex rings.[15] The steps, in a sense, present the simplest possible geometry for a critical phenomenon—a microscopic sphere in the

[14] G. W. Rayfield and F. Reif, Phys. Rev. Letters **11**, 305 (1963); Phys. Rev. **136**, A1194 (1964).

[15] R. P. Feynman, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (Interscience Publishers, Inc., New York, 1955), Vol. I, p. 34; L. Onsager, Nuovo Cimento, Suppl. 2, **6**, 249 (1949). However, for more recent developments, see J. S. Langer and M. E. Fisher, Phys. Rev. Letters **19**, 540 (1967); P. Craig, Phys. Letters **21**, 385 (1966).

bulk liquid—and merit special attention on that account. It is hoped that the present results will clear the way for a more successful treatment of the phenomenon, along the lines, for example, suggested by Di Castro,[3] if, indeed, vortex rings are to provide the answer.

ACKNOWLEDGMENTS

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Monte Carlo Study of a Hydrogenous Plasma near the Ionization Temperature

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Results of a recent Monte Carlo study of a hydrogenous plasma near the ionization temperature show that distribution functions obtained are unusually sensitive to two parameters. The first is the cutoff imposed at small radii on the Coulomb potential between unlike particles, and it becomes necessary to consider quantum-mechanical effects at these radii. The second is the maximum step length Δ through which the particles are allowed to move in the Monte Carlo procedure. It appears that near the ionization temperature the plasma behaves as a mixture of two phases, one ionized, the other un-ionized, and the magnitude chosen for Δ influences which phase dominates.

The problem of obtaining distribution functions for long-range forces has been considered by Broyles, Sahlin, and Carley,[1] and Carley[2] has extended the theory to a classical electron gas. Subsequently, a Monte Carlo (MC) study of a one-component plasma has been completed by Brush, Sahlin, and Teller.[3] In this paper the author presents the results of extending the MC procedure, described in detail by Barker,[4] to a two-component plasma, and particularly considers temperatures in the region where ionization occurs. This region is of considerable interest, but is also the most difficult to deal with from the mathematical standpoint. It is found that for a plasma of density $10^{18}e$/cc at a temperature of 10^5°K, acceptable radial distribution functions are obtained using the MC technique, and below 9×10^3°K the particles become paired, forming the neutral gas. However, in the range 10^4–5×10^4°K, the plasma appears to behave as a mixture of two phases, ionized and un-ionized. Which phase dominates is influenced rather sensitively by a parameter Δ.

As the parameter Δ assumes some importance in the following discussion, the manner in which it arises will be briefly discussed. In MC calculations of this type, a number of particles—16 protons and 16 electrons in this case—are placed in a unit cell. This unit cell is surrounded by a network of identical cells, thus enabling the energy of a configuration to be calculated conveniently as described in Refs. 3 and 4. One particle is displaced a random amount, which can have a maximum value Δ. The energy of the new configuration is calculated, and the MC procedure decides if the move is acceptable or not. Each particle is considered in this manner until the system approaches an equilibrium energy level. The criterion for the choice of Δ is usually based on minimizing the rate of approach of the system to equilibrium; however, as is shown below, the results of this calculation indicate that other considerations should also be taken into account.

Another important choice is the cutoff imposed on the attractive Coulomb potential at short radii. The need for this choice is also encountered when trying to solve integral equations with attractive Coulomb forces present. It can be overcome by treating the close interactions quantum mechanically (QM), and Barker[5] and Storer[6] have independently calculated, with close agreement, effective potentials ϕ_e which should be used when unlike particles approach closer than a certain distance r_J, which depends on temperature. However, the MC calculations were completed previous to the calculation of ϕ_e, and the results are presented in Fig. 1, which shows the unlike radial distribution function g_{MC} obtained from iterations 30 000 to 50 000 with $\Delta=12.5a_0$ (a_0 is the Bohr radius), and using the usual Coulomb potential ϕ_C, but with a constant value below

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[4] A. A. Barker, Aust. J. Physics **18**, 119 (1965).
[5] A. A. Barker, Aust. J. Physics **21** 121 (1968).
[6] R. G. Storer, J. Math. Phys. **9**, 964 (1968).